

PATENT ABSTRACTS OF JAPAN

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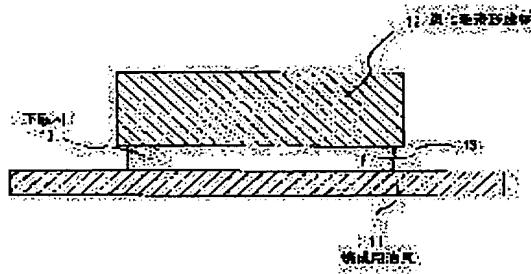
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(54) METHOD FOR FIRING SILICON CARBIDE MOLDED BODY

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a method for firing a silicon carbide molded body, capable of providing a porous silicon carbide member having a sufficiently high flexural strength free from broken piece and pinhole.

SOLUTION: This method for firing a silicon carbide molded body is characterized in that a pillar-shaped silicon carbide molded body which comprises silicon carbide powder, a binder and a dispersion medium liquid and in which a great number of through holes are placed side by side separately by partitions in the longer direction is de-fatted, placed on a burning tool and fired and a space is made between the burning tool and the silicon carbide molded body.



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CLAIMS

[Claim(s)]

[Claim 1] The baking approach of the silicon carbide Plastic solid which is the baking approach of said silicon carbide Plastic solid which lays on the fixture for baking and is calcinated after degreasing the column-like silicon carbide Plastic solid with which many breakthroughs separated the septum and were installed in the longitudinal direction side by side including silicon carbide powder, a binder, and dispersion-medium liquid, and is characterized by preparing space between said fixtures for baking and said silicon carbide Plastic solids.

[Claim 2] The baking approach of a silicon carbide Plastic solid according to claim 1 of preparing space by inserting wooden-clogs material between the fixture for baking, and a silicon carbide Plastic solid.

[Claim 3] The baking approach of the silicon carbide Plastic solid according to claim 2 using a ceramic member as wooden-clogs material.

[Claim 4] A ceramic member is the baking approach of the silicon carbide Plastic solid according to claim 3 which consists of carbon or silicon carbide.

[Claim 5] For a ceramic member, bulk density is 3 0.3g/cm. The baking approach of the silicon carbide Plastic solid according to claim 3 which are the following carbon felt.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the baking approach of a column-like silicon carbide Plastic solid that many breakthroughs were installed in the longitudinal direction side by side.

[0002]

[Description of the Prior Art] It poses a problem that the particulate contained in the exhaust gas discharged by internal combustion engines, such as cars, such as a bus and a truck, and a construction equipment, does damage to an environment or the body recently. By passing a porosity ceramic for this exhaust gas, the ceramic filter which carries out uptake of the particulate in exhaust gas, and purifies exhaust gas is proposed variously.

[0003] Two or more porosity silicon carbide members 90 as shown in drawing 8 band together, and the ceramic filter usually constitutes the ceramic filter 80. Moreover, as this porosity silicon carbide member 90 is shown in drawing 9, many breakthroughs 95 are installed in a longitudinal direction side by side, and the septum 94 which separates breakthrough 95 comrades functions as a filter.

[0004] Namely, the breakthrough 95 formed in the porosity silicon carbide member 90 As shown in drawing 9 (b), the exhaust gas with which either the entry side of exhaust gas or the edge of an outlet side flowed into ***** and the breakthrough 95 of 1 with the filler 96 In case it flows out of other breakthroughs 95 and exhaust gas passes this septum 94 after passing the septum 94 which surely separates a breakthrough 95, a particulate is caught in septum 94 part and exhaust gas is purified. Such a porosity silicon carbide member 90 is extremely excellent in thermal resistance, and since regeneration etc. is easy, it is used for various large-sized cars etc.

[0005] In case such a porosity silicon carbide member is manufactured conventionally, after mixing silicon carbide powder, a binder, and dispersion-medium liquid and preparing the mixed constituent for Plastic solid manufacture first, a silicon carbide Plastic solid is produced by performing extrusion molding of this mixed constituent etc.

[0006] And next, the acquired silicon carbide Plastic solid is dried using a heater etc., it has fixed reinforcement, and the desiccation object of the silicon carbide Plastic solid which can be dealt with easily is manufactured. after this desiccation process -- an obturation process -- carrying out -- the end of the breakthrough of a silicon carbide Plastic solid -- an obturation agent -- being filled up -- one of the breakthroughs -- an end -- **** suggestion **** -- it is made like.

[0007] While heating a silicon carbide Plastic solid at 400-650 degrees C under an oxygen content ambient atmosphere after this desiccation process and an obturation process and volatilizing the solvent in an organic binder component, the cleaning process which carries out decomposition disappearance of the resinous principle is performed, and a porosity silicon carbide member is manufactured through the baking process made to sinter further by heating silicon carbide powder at 2000-2200 degrees C under an inert gas ambient atmosphere.

[0008] And at the conventional baking process, as shown in drawing 7 , after laying silicon carbide Plastic solid 12 on the fixture 11 for baking made from the carbon installed into the crucible (not shown) and carrying this in to a firing furnace, it was calcinating by heating.

[0009] Silicon carbide Plastic solid 12 originates in that manufacture condition, and is about 3% of SiO₂ in silicon carbide powder. Since it contains, it sets at this baking process, and it is silicon carbide Plastic solid 12 to the above SiO₂. The following reaction formula with the carbon which constitutes this SiO gas and the fixture 11 for baking by being sublimated and emitted and that part serving as SiO gas (1);

[0010] $\text{SiO}_2 + 2 \text{C} \rightarrow \text{SiC} + \text{CO} \dots (1)$

[0011] It is alike and the shown reaction advances. consequently -- while the big and rough particle which consists of silicon carbide will be formed in fixture 11 front face for baking and the smooth nature on the front face for baking of a fixture will be lost -- silicon carbide Plastic solid 12 and the fixture 11 for baking -- adhering -- etc. -- it was generated, the porosity silicon carbide member obtained was made to generate a chip 14 and a pinhole, and it had become the cause which the yield reduces.

[0012]

[Problem(s) to be Solved by the Invention] This invention was made in order to solve these problems, it has neither a chip nor a pinhole, and aims at offering the baking approach of a silicon carbide Plastic solid that the porosity silicon carbide member which has flexural strength high enough can be obtained.

[0013]

[Means for Solving the Problem] After the baking approach of the silicon carbide Plastic solid of this invention degreases the column-like silicon carbide Plastic solid with which many breakthroughs separated the septum and were installed in the longitudinal direction side by side including silicon carbide powder, a binder, and dispersion-medium liquid, it is the approach of laying on the fixture for baking and calcinating, and is characterized by preparing space between the above-mentioned fixture for baking, and the above-mentioned silicon carbide Plastic solid.

[0014]

[Embodiment of the Invention] Hereafter, the baking approach of the silicon carbide Plastic solid of this invention is explained as occasion demands, referring to drawing 1. In this invention, the column-like silicon carbide Plastic solid with which many breakthroughs separated the septum and were first installed in the longitudinal direction side by side including silicon carbide powder, a binder, and dispersion-medium liquid is produced.

[0015] Although especially the particle size of the above-mentioned silicon carbide powder is not limited, what combined the powder 100 weight section which has the mean particle diameter which has few contraction is desirable, for example, is about 0.3-50 micrometers, and the powder 5 - 65 weight sections which have the mean particle diameter of about 0.1-1.0 micrometers in the next baking process is desirable.

[0016] Although not limited especially as the above-mentioned binder, methyl cellulose, a carboxymethyl cellulose, hydroxyethyl cellulose, a polyethylene glycol, phenol resin, an epoxy resin, etc. can be mentioned, for example. The loadings of the above-mentioned binder usually have desirable 1 - 10 weight section extent to the silicon carbide powder 100 weight section.

[0017] Although not limited especially as the above-mentioned dispersion-medium liquid, alcohol [, such as an organic solvent; methanol,], such as benzene, water, etc. can be mentioned, for example. Optimum dose combination of the above-mentioned dispersion-medium liquid is carried out so that the viscosity of a mixed constituent may become fixed within the limits.

[0018] After these silicon carbide powder, a binder, dispersion-medium liquid, etc. are mixed by attritor etc., it is fully kneaded by a kneader etc. and the silicon carbide Plastic solid of the shape of a column by which many breakthroughs separated the septum and were installed in the longitudinal direction side by side by the extrusion-molding method etc. is produced. After drying this silicon carbide Plastic solid, an obturation process is performed and ***** of the breakthrough of a silicon carbide Plastic solid is performed.

[0019] Then, the silicon carbide Plastic solid produced by the above-mentioned process is degreased. At the cleaning process of the above-mentioned silicon carbide Plastic solid, after laying the above-mentioned silicon carbide Plastic solid in the fixture for cleaning, it carries in to a cleaning furnace and usually heats at 400-650 degrees C under an oxygen content ambient atmosphere. Thereby, while a binder etc. vaporizes, it decomposes and disappears and only silicon carbide powder remains mostly.

[0020] Next, the degreased silicon carbide Plastic solid is laid on the fixture for baking, and is calcinated. At this baking process, heat the silicon carbide Plastic solid degreased at 2000-2200 degrees C under inert gas ambient atmospheres, such as nitrogen and an argon, silicon carbide powder is made to sinter, and a porosity silicon carbide member is manufactured. In this invention, space is prepared between the above-mentioned fixture for baking, and the above-mentioned silicon carbide Plastic solid in this case. In addition, as the above-mentioned fixture for baking, carbon is usually used.

[0021] Space is prepared for preventing generating a chip and a pinhole in the porosity silicon carbide member

manufactured by the silicon carbide big and rough particle generated when the SiO gas and the fixture for baking made from the above-mentioned carbon generated from the above-mentioned silicon carbide Plastic solid react between the above-mentioned fixture for baking, and the above-mentioned silicon carbide Plastic solid.

[0022] Although not limited especially as an approach of preparing space between the fixture for baking made from the above-mentioned carbon, and the above-mentioned silicon carbide Plastic solid, as shown in drawing 1, the approach of inserting the wooden-clogs material 13 between the fixture 11 for baking and silicon carbide Plastic solid 12 is mentioned, for example.

[0023] Since the thermal resistance which can bear the elevated temperature at the time of baking is required for the wooden-clogs material 13, its ceramic member which has such thermal resistance is desirable.

[0024] When space is formed between the fixture 11 for baking, and silicon carbide Plastic solid 12 using the wooden-clogs material 13 which consists of such a ceramic member, heat transfer to silicon carbide Plastic solid 12 is mainly performed through the fixture 11 for baking which exists under silicon carbide Plastic solid 12, heat conducts it through the wooden-clogs material 13 in the part in which the wooden-clogs material 13 exists, and heat conducts it by thermal radiation in the part in which the wooden-clogs material 13 does not exist.

[0025] And if the heat transfer from the ingredient which exists downward is large, lifting of the temperature of silicon carbide Plastic solid 12 will advance at a rate with early sintering early, on the other hand, if the heat transfer from a lower ingredient is small, lifting of the temperature of silicon carbide Plastic solid 12 will be slow, and sintering will not advance easily.

[0026] Therefore, in order to advance sintering early, as for the above-mentioned ceramic member, what has the comparatively high heat conductivity is desirable, for example, carbon, silicon carbide, aluminium nitride, silicon nitride, etc. are mentioned.

[0027] In this, the above-mentioned silicon carbide does not react at all with SiO gas, but since the porosity silicon carbide member and ingredient which are manufactured are the same, it is desirable, but since this silicon carbide is too hard, it tends to damage the above-mentioned silicon carbide Plastic solid. Therefore, carbon is more desirable when these matters are synthetically taken into consideration. Moreover, although especially the gestalt will not be limited if it is carbon, what finished setting up the thing of the shape of the carbon felt which cannot damage a silicon carbide Plastic solid easily, and which was made blanket-like combining carbon fiber, or yarn is desirable.

[0028] Moreover, the bulk density of the above-mentioned carbon felt is 0.3 g/cm³. It is desirable that it is the following and it is 0.1 g/cm³. It is more desirable that it is the following. It is because do not damage silicon carbide Plastic solid 12, and the touch area of silicon carbide Plastic solid 12 and the carbon felt is small, the omission of the SiO gas which occurred from silicon carbide Plastic solid 12 is also good, although it is inferior to thermal conductivity a little, so the big and rough particle which silicon carbide Plastic solid 12 and the carbon felt react, and consists of silicon carbide is hard to be formed.

[0029] The bulk density of the above-mentioned carbon felt is 0.3 g/cm³. Since the flow of the SiO gas which occurred that it was above from silicon carbide Plastic solid 12 is not good, the big and rough particle which SiO and the carbon felt become easy to react and consists of silicon carbide is easy to be formed, and silicon carbide Plastic solid 12 and the carbon felt adhere, or it becomes easy to generate a chip and a pinhole in silicon carbide Plastic solid 12. In addition, the bulk density of the above-mentioned carbon felt is 0.3 g/cm³. It hurts ** quantity the following and thermal conductivity also becomes low with 0.4 or less W/m·k. Therefore, although it is desirable to make the thickness thin in order to make good heat conduction from the fixture 11 for baking, it is necessary to secure the space for SiO gas escaping. When these things are taken into consideration, the thickness of the carbon felt has about 1-10 desirablemm.

[0030] The thickness of the wooden-clogs material 13 has the desirable range of 1-10mm, when heat conduction from under silicon carbide Plastic solid 12 is taken into consideration. What is necessary is just to adjust concrete thickness suitably in consideration of the thermal conductivity of the wooden-clogs material 13 used actually.

[0031] Although not limited especially as a concrete configuration of the above-mentioned wooden-clogs material, the shape of the field of the stability at the time of laying silicon carbide Plastic solid 12 to the square pole is desirable.

[0032] Moreover, although not limited especially as a touch area of the above-mentioned wooden-clogs material and a silicon carbide Plastic solid, when heat conduction through the above-mentioned wooden-clogs material is taken into consideration, the one where a touch area is larger is desirable. However, in order that SiC may generate by the reaction with SiO gas as described above when carbon is used, the smaller one of the touch area is desirable.

[0033] It may not be limited especially as the number of the wooden-clogs material arranged under a silicon carbide Plastic solid, but two may be laid in the ends of a silicon carbide Plastic solid, and you may lay more than it.

[0034] In addition, at a series of processes of resulting [from a cleaning process] in a baking process, a silicon carbide Plastic solid is carried through wooden-clogs material on the fixture for baking, and it is desirable to perform a cleaning process and a baking process as it is. It is because it can prevent that can perform a cleaning process and a baking process efficiently, and carry, and a silicon carbide Plastic solid gets damaged in a substitute etc.

[0035] Thus, by using the baking approach of the silicon carbide Plastic solid of this invention, there is neither a chip nor a pinhole and a porosity silicon carbide member with high flexural strength can be manufactured.

[0036] Moreover, bulk density is 0.3 g/cm³ as wooden-clogs material especially inserted between the fixture for baking, and a silicon carbide Plastic solid. By using the following carbon felt, there is neither a chip nor a pinhole and a porosity silicon carbide member with high flexural strength can be manufactured suitably.

[0037]

[Example] Although an example is hung up over below and this invention is explained to it in more detail, this invention is not limited only to these examples.

[0038] The mixed constituent of a raw material was prepared by blending the alpha mold silicon carbide powder 70 weight section with example 1 mean particle diameter of 30 micrometers, the beta mold silicon carbide powder 30 weight section with a mean particle diameter of 0.28 micrometers, the methyl cellulose 5 weight section, the dispersant 4 weight section, and the water 20 weight section, and mixing to homogeneity. The extruding press machine was filled up with this mixed constituent, and silicon carbide Plastic solid 23 as shown in drawing 2 by part for extrusion rate/of 2cm was produced. That magnitude was 33mmx33mmx300mm, and the number of breakthroughs 25 was [cm / 31 thickness /of 2 and a septum 24 of this silicon carbide Plastic solid 23] 0.35mm.

[0039] Next, the wooden-clogs material which consists of carbon felt (3mmx5mmx410mm, thermal conductivity:0.24 W/m-K, and bulk density:0.15 g/cm³) was laid on the fixture for baking which consists of porous carbon (Tokai Carbon Co., Ltd. make G100), the silicon carbide Plastic solid dried on this was laid, and the cleaning process was performed by heating at 450 degrees C under the mixed-gas ambient atmosphere of the air and nitrogen which have 5% of oxygen density.

[0040] Next, laying silicon carbide Plastic solid 23 in the above-mentioned fixture for baking, it carried in to baking equipment, and under nitrogen-gas-atmosphere mind, by heating at 2200 degrees C, the silicon carbide Plastic solid was calcinated and the porosity silicon carbide member was manufactured.

[0041] Three-point bending of the obtained porosity silicon carbide member was performed using the assessment approach (1) flexural-strength flexural strength testing machine, and it asked for flexural strength. (2) crystal grain -- ***** -- having had -- the condition of the grain growth of the front face of a porosity silicon carbide member was evaluated with the scanning electron microscope (SEM) photograph.

(3) it evaluated by observing the front face of a chip and a pinhole profit *** porosity silicon carbide member.

[0042] The SEM photograph of the porosity silicon carbide member obtained by this example 1 is shown in drawing 3 . The flexural strength of an average of the sample which abnormality grain growth was not caused and, as for the crystal grain child in the obtained porosity silicon carbide member, sintering advanced as shown in drawing 3 , and it had grown to be even suitable magnitude, and most of a chip or a pinhole was not observed, but was obtained was 52MPa and a value high enough. Since the relation between the thickness of wooden-clogs material and thermal conductivity had agreed with the conditions which calcinate appropriately mostly, this is considered.

[0043] As example 2 wooden-clogs material, the silicon carbide sintered compact was obtained like the example 1 except having used the carbon felt (5mmx5mmx410mm, thermal conductivity 0.24 W/m-K, and bulk density:0.1 g/cm³).

[0044] The SEM photograph of the porosity silicon carbide member obtained by this example 2 is shown in drawing 4. Since sintering was not advancing thoroughly as the crystal grain child in the obtained porosity silicon carbide member showed drawing 4, the flexural strength of an average of the sample which the particle 10 micrometers or less was also observed and was obtained was low a little compared with 47MPa and the case of an example 1. Moreover, most of a chip or a pinhole was not observed. Since it was thicker than the case where the thickness of wooden-clogs material is an example 1, as for this, some is worse than the case where heat conduction to a silicon carbide Plastic solid is an example 1, and it is considered because temperature was not fully going up.

[0045] As example 3 wooden-clogs material, the silicon carbide sintered compact was obtained like the example 1 except having used silicon carbide (2mmx5mmx410mm).

[0046] The SEM photograph of the porosity silicon carbide member obtained by this example 3 is shown in drawing 5. Abnormality grain growth was not caused, and sintering advanced, and the crystal grain child in the obtained porosity silicon carbide member had grown to be even suitable magnitude, as shown in drawing 5. However, some which the big chip generated were in the obtained sample by contacting wooden-clogs material. When flexural strength was measured except for the sample which such a chip generated, average flexural strength was 53MPa and a value high enough. Heat conduction through the wooden-clogs material which this becomes from silicon carbide is considered because it was enough.

[0047] Silicon carbide Plastic solid 23 was produced like example of comparison 1 example 1. This silicon carbide Plastic solid 23 was directly laid on the fixture for cleaning which consists of porous carbon (Tokai Carbon Co., Ltd. make G100), cleaning and baking were performed on an example 1 and these conditions, and the silicon carbide sintered compact was obtained. The SEM photograph of the porosity silicon carbide member obtained in this example 1 of a comparison is shown in drawing 6. As the crystal grain child in the obtained porosity silicon carbide member showed drawing 6, the flexural strength of an average of the sample which abnormality grain growth was observed and was obtained was as low as 42MPa. Moreover, the chip and the pinhole were also observed in the contact surface with the fixture for baking. Since the big and rough particle of silicon carbide was formed in the front face of the fixture for baking, this is considered.

[0048]

[Effect of the Invention] Since the baking approach of the silicon carbide Plastic solid of this invention is as above-mentioned, it has neither a chip nor a pinhole and can obtain the porosity silicon carbide member which has flexural strength high enough.

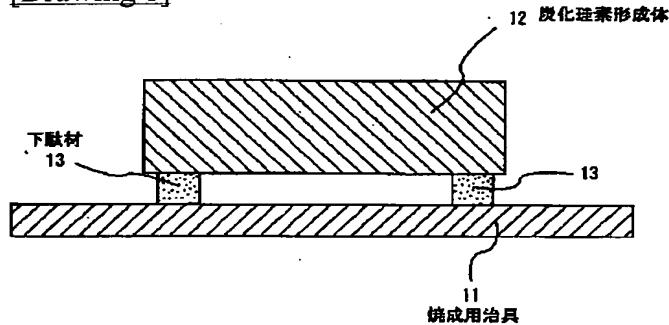
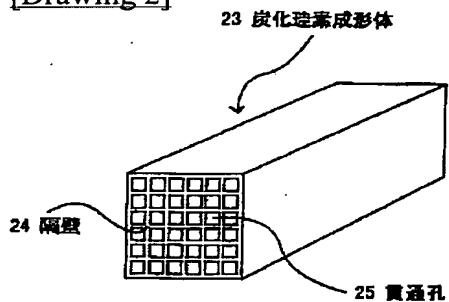
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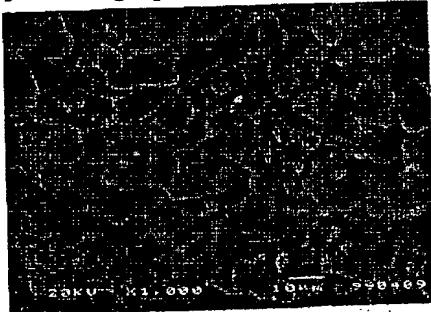
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DRAWINGS

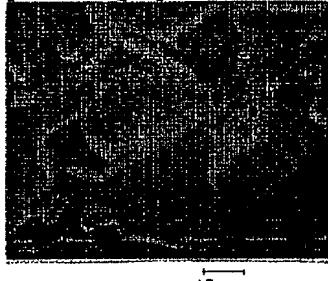
[Drawing 1]**[Drawing 2]****[Drawing 3]****[Drawing 4]**



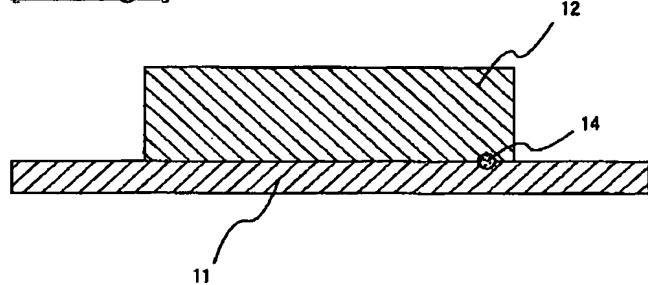
[Drawing 5]



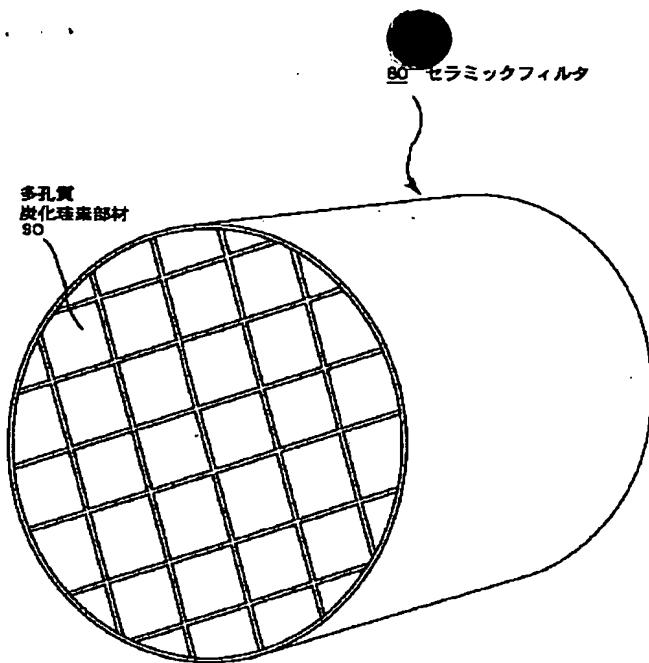
[Drawing 6]

10 μ m

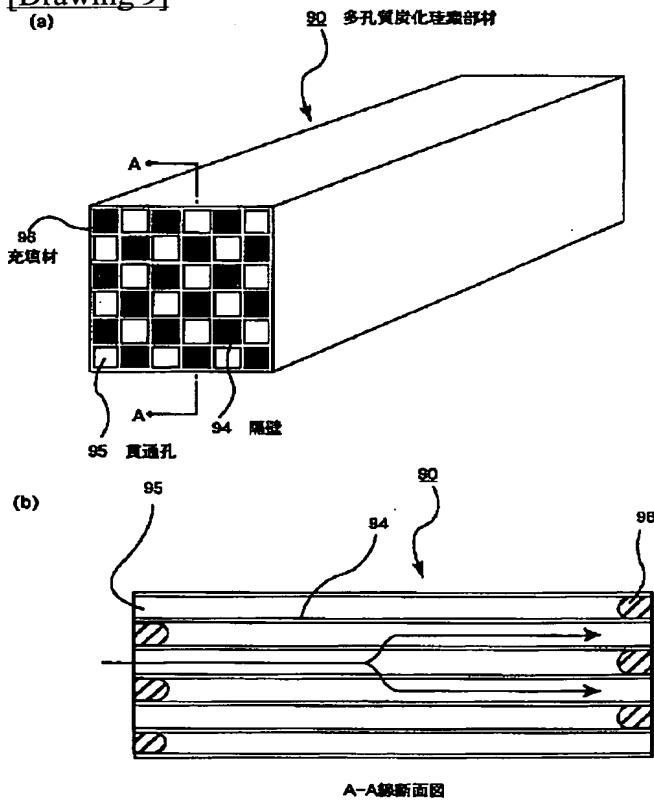
[Drawing 7]



[Drawing 8]



[Drawing 9]



[Translation done.]

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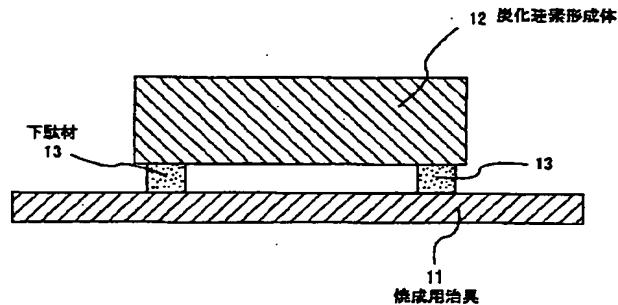
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BD36 BE31

(54) 【発明の名称】炭化珪素成形体の焼成方法

(57) 【要約】

【課題】 欠けやピンホールがなく、充分に高い曲げ強度を有する多孔質炭化珪素部材を得ることができる炭化珪素成形体の焼成方法を提供する。

【解決手段】 炭化珪素粉末とバインダーと分散媒液とを含み、多数の貫通孔が隔壁を隔てて長手方向に並設された柱状の炭化珪素成形体を脱脂した後、焼成用治具上に載置して焼成する上記炭化珪素成形体の焼成方法であって、上記焼成用治具と上記炭化珪素成形体との間に空間を設けることを特徴とする炭化珪素成形体の焼成方法。



【特許請求の範囲】

【請求項1】炭化珪素粉末とバインダーと分散媒液とを含み、多数の貫通孔が隔壁を隔てて長手方向に並設された柱状の炭化珪素成形体を脱脂した後、焼成用治具上に載置して焼成する前記炭化珪素成形体の焼成方法であって、前記焼成用治具と前記炭化珪素成形体との間に空間を設けることを特徴とする炭化珪素成形体の焼成方法。

【請求項2】焼成用治具と炭化珪素成形体との間に下駄材を挿入することにより空間を設ける請求項1記載の炭化珪素成形体の焼成方法。

【請求項3】下駄材として、セラミック部材を用いる請求項2記載の炭化珪素成形体の焼成方法。

【請求項4】セラミック部材は、カーボン又は炭化珪素からなる請求項3記載の炭化珪素成形体の焼成方法。

【請求項5】セラミック部材は、嵩密度が0.3g/cm³以下のカーボンフェルトである請求項3記載の炭化珪素成形体の焼成方法。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】本発明は、多数の貫通孔が長手方向に並設された柱状の炭化珪素成形体の焼成方法に関する。

【0002】

【従来の技術】バス、トラック等の車両や建設機械等の内燃機関から排出される排気ガス中に含有されるパティキュレートが環境や人体に害を及ぼすことが最近問題となっている。この排気ガスを多孔質セラミックを通過させることにより、排気ガス中のパティキュレートを捕集して排気ガスを浄化するセラミックフィルタが種々提案されている。

【0003】セラミックフィルタは、通常、図8に示すような多孔質炭化珪素部材90が複数個結束されてセラミックフィルタ80を構成している。また、この多孔質炭化珪素部材90は、図9に示すように、長手方向に多数の貫通孔95が並設され、貫通孔95同士を隔てる隔壁94がフィルタとして機能するようになっている。

【0004】すなわち、多孔質炭化珪素部材90に形成された貫通孔95は、図9(b)に示すように、排気ガスの入り口側又は出口側の端部のいずれかが充填材96により目封じされ、一の貫通孔95に流入した排気ガスは、必ず貫通孔95を隔てる隔壁94を通過した後、他の貫通孔95から流出するようになっており、排気ガスがこの隔壁94を通過する際、パティキュレートが隔壁94部分で捕捉され、排気ガスが浄化される。このような多孔質炭化珪素部材90は、極めて耐熱性に優れ、再生処理等も容易であるため、種々の大型車両等に使用されている。

【0005】従来、このような多孔質炭化珪素部材を製造する際には、まず、炭化珪素粉末とバインダーと分散

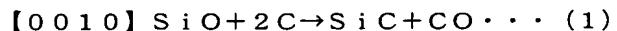
媒液とを混合して成形体製造用の混合組成物を調製した後、この混合組成物の押出成形等を行うことにより、炭化珪素成形体を作製する。

【0006】そして、次に、得られた炭化珪素成形体をヒータ等を用いて乾燥させ、一定の強度を有し、容易に取り扱うことができる炭化珪素成形体の乾燥体を製造する。この乾燥工程の後に封口工程を行って、炭化珪素成形体の貫通孔の一端に封口剤を充填し、貫通孔のどちらか一端が目封じされるようにする。

10 【0007】この乾燥工程および封口工程の後、炭化珪素成形体を酸素含有雰囲気下において、400～650℃に加熱し、有機バインダー成分中の溶剤を揮発するとともに、樹脂成分を分解消失させる脱脂工程を行い、さらに、炭化珪素粉末を不活性ガス雰囲気下、2000～2200℃に加熱することにより焼結させる焼成工程を経て、多孔質炭化珪素部材が製造される。

【0008】そして、従来の焼成工程では、図7に示すように、るつぼ(図示せず)中に設置されたカーボン製の焼成用治具11上に炭化珪素成形体12を載置し、これを焼成炉に搬入した後、加熱することにより焼成していた。

【0009】炭化珪素成形体12は、その製造条件に起因して炭化珪素粉末中に約3%程度のSiO₂を含んでいるため、この焼成工程において、炭化珪素成形体12から上記SiO₂が昇華して放出され、その一部がSiOガスとなり、このSiOガスと焼成用治具11を構成する炭素との下記反応式(1)：



【0010】に示す反応が進行する。その結果、炭化珪素からなる粗大粒子が焼成用治具11表面に形成され、焼成用治具表面の平滑性が失われてしまうとともに、炭化珪素成形体12と焼成用治具11とのくつき等が生じ、得られる多孔質炭化珪素部材に欠け14やピンホールを発生させ、歩留まりが低減する原因となっていた。

【0012】

【発明が解決しようとする課題】本発明は、これらの問題を解決するためになされたもので、欠けやピンホールがなく、充分に高い曲げ強度を有する多孔質炭化珪素部材を得ることができる炭化珪素成形体の焼成方法を提供することを目的とするものである。

【0013】

【課題を解決するための手段】本発明の炭化珪素成形体の焼成方法は、炭化珪素粉末とバインダーと分散媒液とを含み、多数の貫通孔が隔壁を隔てて長手方向に並設された柱状の炭化珪素成形体を脱脂した後、焼成用治具上に載置して焼成する方法であって、上記焼成用治具と上記炭化珪素成形体との間に空間を設けることを特徴とする。

【0014】

【発明の実施の形態】以下、本発明の炭化珪素成形体の

焼成方法について、必要により、図1を参照しながら説明する。本発明では、初めに、炭化珪素粉末とバインダーと分散媒液とを含み、多数の貫通孔が隔壁を隔てて長手方向に並設された柱状の炭化珪素成形体を作製する。

【0015】上記炭化珪素粉末の粒径は特に限定されないが、後の焼成過程で収縮が少ないものが好ましく、例えば、0.3～50μm程度の平均粒子径を有する粉末100重量部と0.1～1.0μm程度の平均粒子径を有する粉末5～65重量部とを組み合わせたものが好ましい。

【0016】上記バインダーとしては特に限定されないが、例えば、メチルセルロース、カルボキシメチルセルロース、ヒドロキシエチルセルロース、ポリエチレンリコール、フェノール樹脂、エポキシ樹脂等を挙げることができる。上記バインダーの配合量は、通常、炭化珪素粉末100重量部に対して、1～10重量部程度が好ましい。

【0017】上記分散媒液としては特に限定されないが、例えば、ベンゼン等の有機溶媒；メタノール等のアルコール、水等を挙げができる。上記分散媒液は、混合組成物の粘度が一定範囲内となるように、適量配合される。

【0018】これら炭化珪素粉末とバインダーと分散媒液等とは、アトライター等で混合された後、ニーダー等で充分に混練され、押し出し成形法等により、多数の貫通孔が隔壁を隔てて長手方向に並設された柱状の炭化珪素成形体が作製される。この炭化珪素成形体の乾燥を行った後、封口工程を行い、炭化珪素成形体の貫通孔の目封じを行う。

【0019】この後、上記工程により作製された炭化珪素成形体の脱脂を行う。上記炭化珪素成形体の脱脂工程では、通常、上記炭化珪素成形体を脱脂用治具に載置した後、脱脂炉に搬入し、酸素含有雰囲気下、400～650℃に加熱する。これにより、バインダー等が揮散するとともに、分解、消失し、ほぼ炭化珪素粉末のみが残留する。

【0020】次に、脱脂した炭化珪素成形体を、焼成用治具上に載置して焼成する。この焼成工程では、窒素、アルゴン等の不活性ガス雰囲気下、2000～2200℃で脱脂した炭化珪素成形体を加熱し、炭化珪素粉末を焼結させて多孔質炭化珪素部材を製造する。本発明では、この際に、上記焼成用治具と上記炭化珪素成形体との間に空間を設ける。なお、上記焼成用治具としては、通常、カーボンが用いられる。

【0021】上記焼成用治具と上記炭化珪素成形体との間に空間を設けるのは、上記炭化珪素成形体から発生するSiOガスと上記カーボン製の焼成用治具とが反応することにより生成する炭化珪素粗大粒子により、製造される多孔質炭化珪素部材に欠けやピンホールを発生するのを防止するためである。

【0022】上記カーボン製の焼成用治具と上記炭化珪素成形体との間に空間を設ける方法としては特に限定されないが、例えば、図1に示したように、焼成用治具11と炭化珪素成形体12との間に下駄材13を挿入する方法が挙げられる。

【0023】下駄材13は、焼成時の高温に耐え得る耐熱性が必要であることから、そのような耐熱性を有するセラミック部材が好ましい。

【0024】このようなセラミック部材からなる下駄材13を用いて焼成用治具11と炭化珪素成形体12との間に空間を形成した際、炭化珪素成形体12への伝熱は、主に、炭化珪素成形体12の下に存在する焼成用治具11を介して行われ、下駄材13が存在する部分では、下駄材13を介して熱が伝導し、下駄材13が存在しない部分では、熱放射により熱が伝導する。

【0025】そして、下に存在する材料からの伝熱が大きければ、炭化珪素成形体12の温度の上昇が早く、焼結が早い速度で進行し、一方、下の材料からの伝熱が小さければ、炭化珪素成形体12の温度の上昇が遅く、焼結がなかなか進行しない。

【0026】従って、焼結を早く進行させるためには、上記セラミック部材は、比較的熱伝導率の高いものが好ましく、例えば、カーボン、炭化珪素、窒化アルミニウム、窒化ケイ素等が挙げられる。

【0027】このなかで上記炭化珪素は、SiOガスと全く反応せず、製造される多孔質炭化珪素部材と材料が同じであるため好ましいが、この炭化珪素は硬すぎるために上記炭化珪素成形体を傷付けやすい。従って、これらの事項を総合的に勘案すると、カーボンがより好ましい。また、カーボンであれば、その形態は特に限定されないが、炭化珪素成形体を傷付けにくい、カーボン繊維を組み合わせて布状にしたカーボンフェルトや糸状のものを組み上げたもの等が好ましい。

【0028】また、上記カーボンフェルトの嵩密度は、0.3g/cm³以下であることが好ましく、0.1g/cm³以下であることがより好ましい。熱伝導性には若干劣るもの、炭化珪素成形体12を傷付けることがなく、また、炭化珪素成形体12とカーボンフェルトとの接触面積が小さく、炭化珪素成形体12から発生したSiOガスの抜けも良いので、炭化珪素成形体12とカーボンフェルトとが反応して炭化珪素からなる粗大粒子が形成されにくいからである。

【0029】上記カーボンフェルトの嵩密度が0.3g/cm³以上であると、炭化珪素成形体12から発生したSiOガスの流れがよくないため、SiOとカーボンフェルトとが反応しやすくなり、炭化珪素からなる粗大粒子が形成されやすく、炭化珪素成形体12とカーボンフェルトとがくっついたり、炭化珪素成形体12に欠けやピンホールが発生しやすくなる。なお、上記カーボンフェルトの嵩密度は、0.3g/cm³以下と嵩高いた

め、熱伝導率も $0.4\text{ W/m}\cdot\text{K}$ 以下と低くなる。従つて、焼成用治具11からの熱伝導を良好にするためには、その厚さを薄くすることが望ましいが、SiOガスが逃散するための空間は確保しておく必要がある。これらのことから勘案すると、カーボンフェルトの厚さは、1~10mm程度が望ましい。

【0030】下駄材13の厚さは、炭化珪素成形体12の下からの熱伝導を考慮すると、1~10mmの範囲が好ましい。具体的な厚さは、実際に用いる下駄材13の熱伝導率を考慮して適宜調整すればよい。

【0031】上記下駄材の具体的な形状としては特に限定されないが、炭化珪素成形体12を載置した際の安定性の面から四角柱状が好ましい。

【0032】また、上記下駄材と炭化珪素成形体との接触面積としては特に限定されないが、上記下駄材を介した熱伝導を考慮すると接触面積が大きい方が好ましい。しかし、カーボンを使用した場合には、上記したように、SiOガスとの反応によりSiCが生成するため、その接触面積は小さい方が好ましい。

【0033】炭化珪素成形体の下に配置する下駄材の数としては特に限定されず、炭化珪素成形体の両端に2本載置しても良いし、それ以上載置しても良い。

【0034】なお、脱脂工程から焼成工程に至る一連の工程では、焼成用治具上に下駄材を介して炭化珪素成形体を載せ、そのまま、脱脂工程及び焼成工程を行うことが好ましい。脱脂工程及び焼成工程を効率的に行うことができ、また、載せ代え等において、炭化珪素成形体が傷つくのを防止することができるからである。

【0035】このように本発明の炭化珪素成形体の焼成方法を用いることにより、欠けやピンホールがなく、曲げ強度の高い多孔質炭化珪素部材を製造することができる。

【0036】また、特に、焼成用治具と炭化珪素成形体との間に挿入する下駄材として嵩密度が 0.3 g/cm^3 以下のカーボンフェルトを用いることにより、欠けやピンホールがなく、曲げ強度の高い多孔質炭化珪素部材を好適に製造することができる。

【0037】

【実施例】以下に実施例を掲げて本発明を更に詳しく説明するが、本発明はこれら実施例のみに限定されるものではない。

【0038】実施例1

平均粒子径 $30\mu\text{m}$ の α 型炭化珪素粉末70重量部、平均粒子径 $0.28\mu\text{m}$ の β 型炭化珪素粉末30重量部、メチルセルロース5重量部、分散剤4重量部、水20重量部を配合して均一に混合することにより、原料の混合組成物を調製した。この混合組成物を押出成形機に充填し、押出速度 $2\text{ cm}/\text{分}$ にて図2に示すような炭化珪素成形体23を作製した。この炭化珪素成形体23は、その大きさが $33\text{ mm}\times 33\text{ mm}\times 300\text{ mm}$ で、貫通孔

25の数が31個/ cm^2 、隔壁24の厚さが 0.35 mm であった。

【0039】次に、ポーラスカーボン（東海カーボン社製 G100）からなる焼成用治具上に、カーボンフェルト（ $3\text{ mm}\times 5\text{ mm}\times 410\text{ mm}$ 、熱伝導率： $0.24\text{ W/m}\cdot\text{K}$ 、嵩密度： 0.15 g/cm^3 ）からなる下駄材を載置し、この上に乾燥した炭化珪素成形体を載置して、5%の酸素濃度を有する空気と窒素との混合ガス雰囲気下、 450°C で加熱することにより脱脂工程を行った。

【0040】次に、炭化珪素成形体23を上記焼成用治具に載置したまま、焼成装置に搬入し、窒素ガス雰囲気下、 2200°C に加熱することにより炭化珪素成形体の焼成を行い、多孔質炭化珪素部材を製造した。

【0041】評価方法

(1) 曲げ強度

曲げ強度試験機を用い、得られた多孔質炭化珪素部材の3点曲げを行い、曲げ強度を求めた。

(2) 結晶粒成長

得られた多孔質炭化珪素部材の表面の結晶粒成長の状態を、走査型電子顕微鏡（SEM）写真により評価した。

(3) 欠け、ピンホール

得られた多孔質炭化珪素部材の表面を観察することにより評価した。

【0042】本実施例1で得られた多孔質炭化珪素部材のSEM写真を図3に示す。得られた多孔質炭化珪素部材中の結晶粒子は、図3に示したように、異常粒成長を起こしておらず、かつ、焼結が進行して適當な大きさにまで成長しており、欠けやピンホールは殆ど観察されず、得られたサンプルの平均の曲げ強度は、 52 MPa と充分に高い値であった。これは、下駄材の厚さと熱伝導率との関係が、焼成を適切に行う条件とほぼ合致していたためと考えられる。

【0043】実施例2

下駄材として、カーボンフェルト（ $5\text{ mm}\times 5\text{ mm}\times 410\text{ mm}$ 、熱伝導率 $0.24\text{ W/m}\cdot\text{K}$ 、嵩密度： 0.1 g/cm^3 ）を用いた以外は、実施例1と同様にして炭化珪素焼結体を得た。

【0044】本実施例2で得られた多孔質炭化珪素部材のSEM写真を図4に示す。得られた多孔質炭化珪素部材中の結晶粒子は、図4に示したように、焼結が完全には進行していないため、 $10\mu\text{m}$ 以下の粒子も観察され、得られたサンプルの平均の曲げ強度は、 47 MPa と実施例1の場合と比べると若干低かった。また、欠けやピンホールは殆ど観察されなかった。これは、下駄材の厚さが実施例1の場合より厚かったため、炭化珪素成形体への熱伝導が実施例1の場合より若干悪く、温度が充分に上がっていなかったためと考えられる。

【0045】実施例3

下駄材として、炭化珪素（ $2\text{ mm}\times 5\text{ mm}\times 410\text{ m}$

m) を用いた以外は、実施例 1 と同様にして炭化珪素焼結体を得た。

【0046】本実施例 3 で得られた多孔質炭化珪素部材の SEM 写真を図 5 に示す。得られた多孔質炭化珪素部材中の結晶粒子は、図 5 に示したように、異常粒成長を起こしておらず、かつ、焼結が進行して適当な大きさにまで成長していた。ただし、得られたサンプルのなかには、下駄材と接触することにより大きな欠けが発生したものもあった。このような欠けが発生したサンプルを除いて曲げ強度を測定したところ、平均の曲げ強度は、5 3 MPa と充分に高い値であった。これは、炭化珪素からなる下駄材を介した熱伝導は充分であったためと考えられる。

【0047】比較例 1

実施例 1 と同様にして炭化珪素成形体 23 を作製した。この炭化珪素成形体 23 を、ポーラスカーボン（東海カーボン社製 G100）からなる脱脂用治具上に直接載置して、実施例 1 と同条件で脱脂および焼成を行い炭化珪素焼結体を得た。本比較例 1 で得られた多孔質炭化珪素部材の SEM 写真を図 6 に示す。得られた多孔質炭化珪素部材中の結晶粒子は、図 6 に示したように、異常粒成長が観察され、得られたサンプルの平均の曲げ強度は、42 MPa と低かった。また、焼成用治具との接触面に欠けやピンホールも観察された。これは、焼成用治具の表面に炭化珪素の粗大粒子が形成されたためと考えられる。

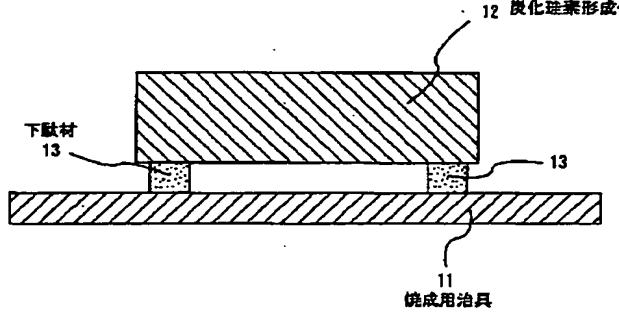
【0048】

【発明の効果】本発明の炭化珪素成形体の焼成方法は、上述の通りであるので、欠けやピンホール等がなく、充分に高い曲げ強度を有する多孔質炭化珪素部材を得るこ

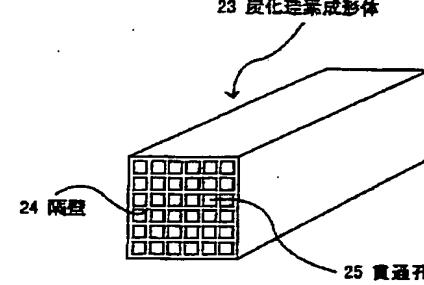
とができる。

【図面の簡単な説明】

【図 1】



【図 2】



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【図 1】本発明の炭化珪素成形体の焼成方法の一例を模式的に示す縦断面図である。

【図 2】本発明の炭化珪素成形体の焼成方法において使用する炭化珪素成形体を模式的に示す斜視図である。

【図 3】実施例 1 で得られた多孔質炭化珪素部材の表面の SEM 写真である。

【図 4】実施例 2 で得られた多孔質炭化珪素部材の表面の SEM 写真である。

【図 5】実施例 3 で得られた多孔質炭化珪素部材の表面の SEM 写真である。

【図 6】比較例 1 で得られた多孔質炭化珪素部材の表面の SEM 写真である。

【図 7】従来の炭化珪素成形体の焼成方法を模式的に示す縦断面図である。

【図 8】セラミックフィルタを模式的に示す斜視図である。

【図 9】(a) は、セラミックフィルタを構成する多孔質炭化珪素部材を模式的に示す斜視図であり、(b) は、(a) に示す多孔質炭化珪素部材の A-A 線断面図である。

【符号の説明】

- 1 1 焼成用治具
- 1 2 炭化珪素成形体
- 1 3 下駄材
- 1 4 欠け
- 2 3 炭化珪素成形体
- 2 4、9 4 隔壁
- 2 5、9 5 貫通孔
- 8 0 セラミックフィルタ
- 9 0 多孔質炭化珪素部材
- 9 6 充填材

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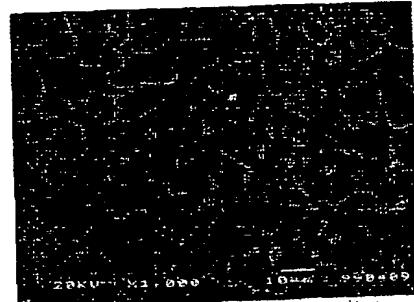
【図3】



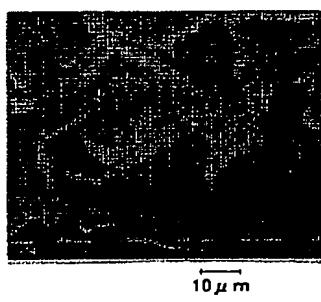
【図4】



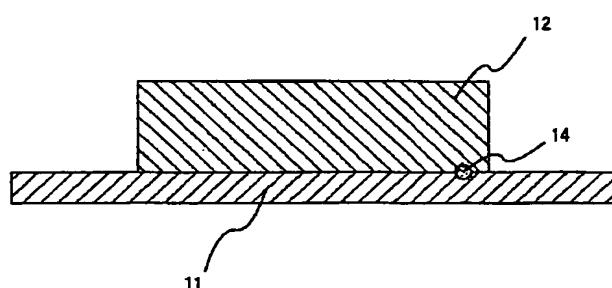
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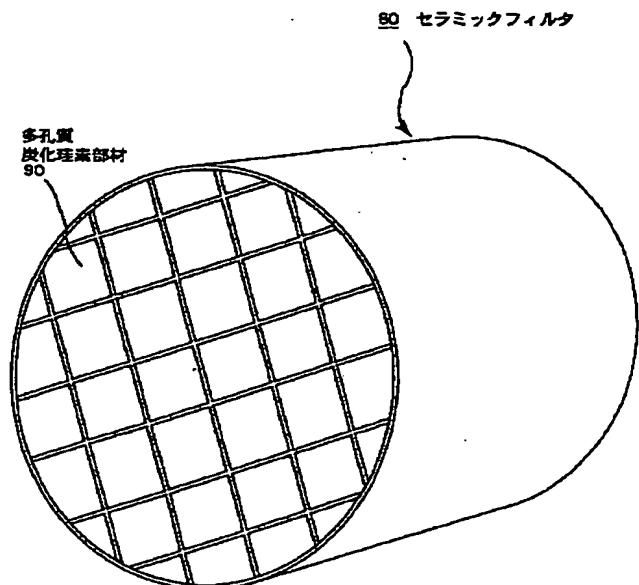
【図6】



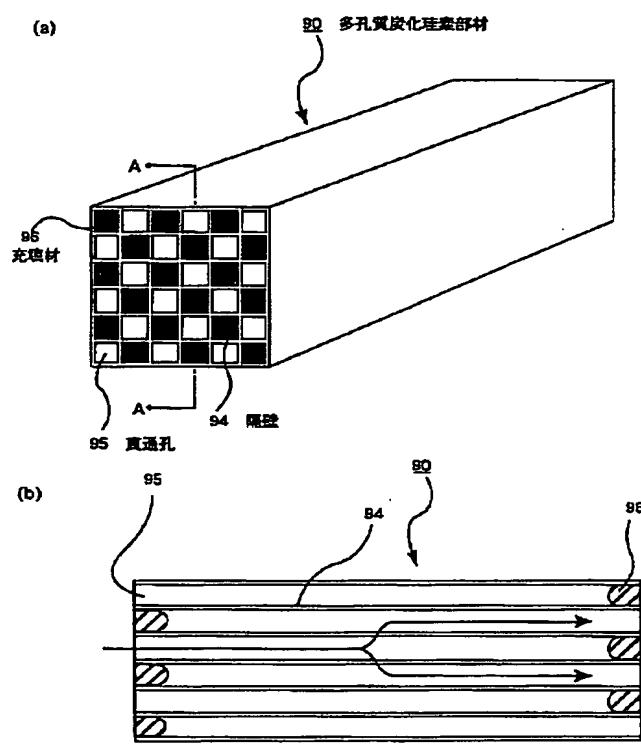
【図7】



【図8】



【図9】



A-A線断面図

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